/N:

NASA Contractor Report 189174

Characterization of Polybenzimidazole (PBI) Film at High Temperatures

Ahmad N. Hammoud Sverdrup Technology, Inc. Lewis Research Center Group Brook Park, Ohio

and

J.L. Suthar SUNY at Buffalo Space Power Insulation Institute Buffalo, New York

April 1992

Prepared for Lewis Research Center Under Contract NAS3-25266



N92-23538

A. N. Hammoud Sverdrup Technology, Inc. NASA Lewis Research Center Group Cleveland, OH 44135

J. L. Suthar SUNY at Buffalo Space Power Insulation Institute Buffalo, NY 14260

ABSTRACT

Polybenzimidazole, a linear thermoplastic polymer with excellent thermal stability and strength retention over a wide range of temperature, was evaluated for its potential use as the main dielectric in high temperature capacitors. The film was characterized in terms of its dielectric properties in a frequency range of 50 Hz to 100 kHz. These properties, which included the dielectric constant and dielectric loss, were also obtained in a temperature range from 20°C to 300°C with an electrical stress of 60 Hz, 50 V/mil present. The ac and dc breakdown voltages of silicone oil-impregnated films as a function of temperature were also determined. The results obtained indicate that while the film remained relatively stable up to 200°C, it exhibited an increase in its dielectric properties as the temperature was raised to 300°C. It was also found that conditioning of the film by heat-treatment at 60°C for 6 hours tend to improve its dielectric and breakdown properties. The results are discussed and conclusions made concerning the suitability of the film as high temperature capacitor dielectric.

INTRODUCTION

Dielectric and insulating materials are used extensively in many electrical systems and components such as high voltage capacitors, power switches, and energy storage and transport devices. These components and devices are often required to operate reliably in harsh environments where stresses of different kinds and intensities are encountered. High temperature constitutes one of such stresses which exists in applications such as nuclear reactors, well-logging fields and space-based systems. The dielectric materials employed in these fields must therefore operate reliably and be capable of withstanding the high temperature exposure.

The currently emerging demand for space-based power systems and components is toward higher energy densities and larger power levels. For instance, future plans for many space exploration missions call for tremendous increase in the power capability to a magnitude of the order of megawatts [1]. Reducing size and weight, increasing the packaging density, as well as improving efficiency and reliability are other issues that are of major concern to space systems. These requirements will certainly result in raising the operating temperature of the device/system concerned [2]. Improvement in the currently available dielectrics and the identification of new materials, capable of providing reliable and efficient operation at high temperatures, thus play an important role for the technological needs in space-based enterprises to be met.

Polybenzimidazole (PBI), a linear thermoplastic polymer, was evaluated for use as high temperature capacitor dielectric. The material was characterized with and without heat treatment in terms of its dielectric properties in a frequency range of 50 Hz to 100 kHz. These properties, which included the dielectric constant and dielectric loss, were also obtained in a temperature range from 20°C to 300°C with an electrical stress of 60 Hz, 50 V/mil present. The ac and dc breakdown voltages of silicone oil-impregnated films as a function of temperature were also determined. In this paper, the experimental procedures and the results obtained are discussed.

EXPERIMENTAL PROCEDURE

PBI films of 1.5 mil thickness, manufactured by Hoechst Celanese, were used in these investigations. The PBI film is a linear thermoplastic polymer which has excellent thermal stability and strength retention over a wide range of temperature [3]. It is chemically stable and is used as reinforcement of high performance composites, filament winding and structural applications. Some of the properties of the film are given in Table

Table I. Properties of PBI film [3].

Service temperature (°C)	>300
Shrinkage (%) @ 315°C	3
Density (g/cc)	1.2-1.4
Dielectric constant	4.4-16.2
Dissipation factor (x10 ⁻²)	2.4-57
Dielectric strength (kV/mil)	1014-1016
Volume resistivity (ohm.cm)	1014-1016
Surface resistivity (ohm/sq.)	5x10 ¹⁰

The experiments carried out were performed on as-received (control C) as well as on heattreated (HT) samples. Heat treatment was done by heating the material in an oven at a temperature of 60°C for a time duration of 6 hours. A capacitance measurement system (General Radio Precision Capacitance System 1621) together with a set of concentric ring brass electrodes were used in the measurement of the dielectric constant and the dissipation factor of the samples at room temperature in a frequency range of 50 Hz to 100 kHz. These properties were further characterized in a temperature range of 20°C to 300°C with an applied electrical stress of 60 Hz, 50 V/mil using Tettex Instruments, Type 2821 Capacitance System and Type 2914 Dielectric Test Cell.

A Hipotronics ac Dielectric Test Set (Model 7100 - 20 A) and a Universal Voltronics dc power supply (Model BAM-32-1.5) were employed in the breakdown voltage measurements. Silicone fluid 210 H, supplied by Dow Corning, served as the impregnant. A temperature controller was utilized to obtain proper test temperatures within $\pm 2^{\circ}$ C. During testing, the specimen was held between two cylindrical stainless steel electrodes of 0.25 inch diameter (ASTM D-149) and the voltage was raised at a rate of 500 V/s until breakdown occurred. The values reported are the average of at least seven measurements.

RESULTS AND DISCUSSIONS

The dielectric constant and the dissipation factor at room temperature as a function of frequency are shown in Figures 1 and 2, respectively. These properties of the film were obtained in the frequency range of 50 Hz to 100 kHz for both the as-received as well as those of heat-treated sample at 60°C for 6 hours. It can be clearly seen that both properties exhibit significant decrease upon heat treatment. For example, at any given frequency, the decrease in the dielectric constant amounted to about as much as 50% of the original value. Similarly, the value of the dissipation factor decreased sharply after the film was heat-treated. It is believed that the reduction in the values of the dielectric properties after heat treatment is due to the removal of any moisture present and possibly to some thermally-induced molecular agitation phenomenon in the polymer.

Figure 3 shows the variation in the dielectric constant of the heat-treated samples as a function of temperature while electrically stressed at 60 Hz, 50 V/mil. It is evident that the dielectric constant remained relatively stable with temperature up to 200°C. As the test temperature was increased further, the value of the dielectric constant, however, started to exhibit an increase. A similar trend was observed for the effect of temperature on the dissipation factor of the heat-treated PBI film, as shown in Figure 4. It is important to note that the polymer film maintained its physical integrity at temperatures as high as 300°C and in the presence of the electrical stress.

A comparison of the ac and dc breakdown voltages at room temperature of control and heat-treated samples is given in Table II. Also listed are the data for both dry as well as for silicone fluid-impregnated samples. In general, the breakdown strength increased slightly after heat treatment. Once again, this might have been due to the removal of trapped moisture upon heating the material. Table II also shows that the breakdown voltages of the impregnated samples are much higher than their dry counterpart. This happens because the impregnant, which has higher dielectric strength than air, penetrates the material and fills up the microvoids and gas cavities as these are usually considered as primary sites for breakdown initiation.

The ac and dc breakdown strengths of impregnated PBI films before and after heat

treatment as a function of temperature are shown in Figures 5 and 6, respectively. It can be seen that for either case, ac or dc, the dielectric strength reduces slightly with increase in temperature. This reduction in the breakdown voltage can be attributed to the softening of the polymer film when exposed to high temperatures. It can be also noted that the heat-treated samples displayed, at any given temperature, higher dielectric strengths than those of the untreated film. This is due to the fact that in addition to removing any trapped moisture in the film, the application of heat would have facilitated and lead to better impregnation of the material.

CONCLUSION

The results obtained in this work indicate that the PBI film remained relatively stable when exposed to temperatures as high as 200°C. At higher temperatures, the film, however, exhibited an increase in its dielectric properties. Its breakdown behavior displayed weak dependence on temperature as both the ac and dc dielectric strengths exhibited slight decrease with increase in temperature. It is believed that improvement in the manufacturing and processing of the film can possibly result in good electrical and other properties that are more stable with temperature. Further research and experimental studies are required to better and fully characterize this and other materials for potential use as high temperature capacitor dielectrics.

ACKNOWLEDGEMENTS

This work was supported by NASA Lewis Research Center, Contract # NAS3-25266, Task Order # 5423-01, "High Temperature Dielectrics." The authors thank J. E. Ramirez and E. J. Powers (Hoechst Celanese Co.) for supplying the film and I. Myers and S. Domitz (NASA LeRC) for their technical contribution.

REFERENCES

- [1] R. W. Bercaw, "Toward an Electrical Power Utility for Space Exploration," Proceedings of the European Space Power Conference, Madrid, Spain, October 2-6, 1989.
- [2] E. Sugimoto, "Applications of Polyimide Films to the Electrical and Electronic Industries in Japan," IEEE Electrical Insulation Magazine, Vol. 5, No. 1, 1989.
- [3] E. J. Powers and G. A. Serad, "History and Development of Polybenzimidazoles," in <u>High Performance Polymers: Their Origin and Development</u>, R. B. Seymour and G. S. Krishenbaum, editors, Elsevier Pub., 1986.

Table II. Effect of heat treatment on the dielectric strengths of dry and impregnated PBI films.

	ac strength, kV		dc strength, kV	
sample	dry	imp	dry	imp
as-received	5.69	9.36	8.52	9.13
heat-treated	6.36	9.40	9.75	10.26

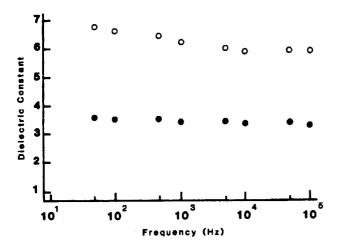


Figure 1. Dielectric constant versus frequency for control(0) and heat-treated(e) samples.

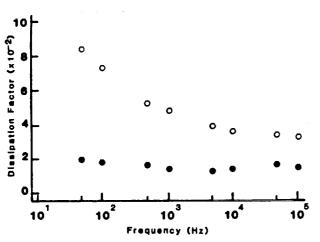


Figure 2. Dissipation factor versus frequency for control(0) and heat-treated(0) samples.

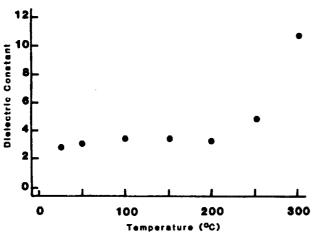


Figure 3. Dielectric constant of heat-treated samples versus temperature while stressed at 60 Hz, 50 V/mll.

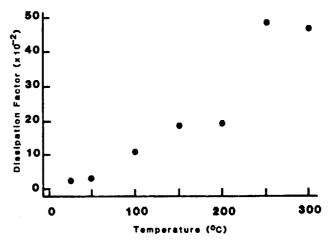


Figure 4. Dissipation factor of heat-treated samples versus temperature while stressed at 60 Hz, 50 V/mil.

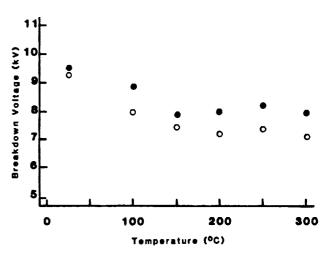


Figure 5. Dependence of ac dielectric strength on temperature. (D:control@:heat_treated)

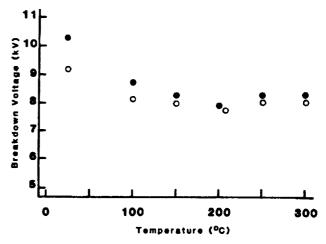


Figure 6. Dependence of dc dielectric strength on temperature. (O:control@:heat-treated)

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Artington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

Davis Highway, Suite 1204, Arlington, VA 223					
1. AGENCY USE ONLY (Leave blank)		į			
	April 1992		I Contractor Report		
4. TITLE AND SUBTITLE	' '		FUNDING NUMBERS		
Characterization of Polyber	zimidazole (PBI) Film at High	Temperatures			
			WU-506-41-41		
6. AUTHOR(S)		· · · · · · · · · · · · · · · · · · ·	C-NAS3-25266		
Ahmad N. Hammoud and J	I Cuthor				
Annad N. Hammoud and J	.L. Sutilai				
7. PERFORMING ORGANIZATION N	AME(S) AND ADDRESS(ES)	8.	PERFORMING ORGANIZATION		
	(0)	•	REPORT NUMBER		
Sverdrup Technology, Inc.	Sverdrup Technology, Inc.				
Lewis Research Center Gro	up		E-6957		
2001 Aerospace Parkway			_ 3,2,		
Brook Park, Ohio 44142					
9. SPONSORING/MONITORING AGE	NCY NAMES(S) AND ADDRESS(ES	10	SPONSORING/MONITORING		
			AGENCY REPORT NUMBER		
National Aeronautics and S	pace Administration				
Lewis Research Center	101		NASA CR-189174		
Cleveland, Ohio 44135-31	.91				
11. SUPPLEMENTARY NOTES		<u>. </u>			
	, Power Technology Division, N	JASA Lewis Research Cen	ter (216) 433–6116		
	drup Technology, Inc.; and J.L.		• • •		
Institute, Buffalo, New York					
12a. DISTRIBUTION/AVAILABILITY S		12	b. DISTRIBUTION CODE		
,					
Unclassified - Unlimited					
Subject Category 33					
13. ABSTRACT (Maximum 200 words					
	thermoplastic polymer with ex-				
	wide range of temperature, was evaluated for its potential use as the main dielectric in high temperature capacitors.				
	The film was characterized in terms of its dielectric properties in a frequency range of 50 Hz to 100 kHz. These				
	he dielectric constant and dielec				
	etrical stress of 60 Hz, 50 V/mil				
oil-impregnated films as a function of temperature were also determined. The results obtained indicate that while the film remained relatively stable up to 200 °C, it exhibited an increase in its dielectric properties as the tempera-					
	t was also found that conditioni breakdown properties. The res				
	h temperature capacitor dielectr		rusions made concerning the		
sultability of the film as high	remperature capacitor dielecti	ю.			
•					
14. SUBJECT TERMS			15. NUMBER OF PAGES		
Insulation dielectrics; Capacitors; High temperature		4			
•			16. PRICE CODE A02		
17. SECURITY CLASSIFICATION	18. SECURITY CLASSIFICATION	19. SECURITY CLASSIFICATIO			
OF REPORT	OF THIS PAGE	OF ABSTRACT			
Unclassified	Unclassified	Unclassified			